

Novel Ground States in Epitaxially-Grown 2D Superconductor LaSb₂



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Introduction

- Recent discoveries^{2,3} in the coupling between electronic and structural characteristics of the family of Rare earth diantimonide (*RSb*₂) stacked intermetallic structures has prompted research in the coupling by utilizing structural change through the application of hydrostatic pressure and temperature.
- Members of this *RSb*₂ family exhibit desirable electronic characteristics in areas of active research, such as square-nets of ions and superconductivity.
- LaSb₂, a non-magnetic member of the *RSb*₂ family, is known to exhibit structural change under applied hydrostatic pressure in the bulk crystal². In the research described by this poster and accompanying manuscript¹, LaSb₂ was structurally manipulated by growing the material in the thin-film limit under epitaxial strain.
- The results of this research revealed a novel ground state for LaSb₂, differing from the well-studied bulk crystal (SG #64 Cmca) by a structural change from Orthorhombic to Monoclinic, synthesizing in SG #12 A 2/m.
- Additionally, ab-initio calculations predict a second hitherto unknown polymorph of the system.

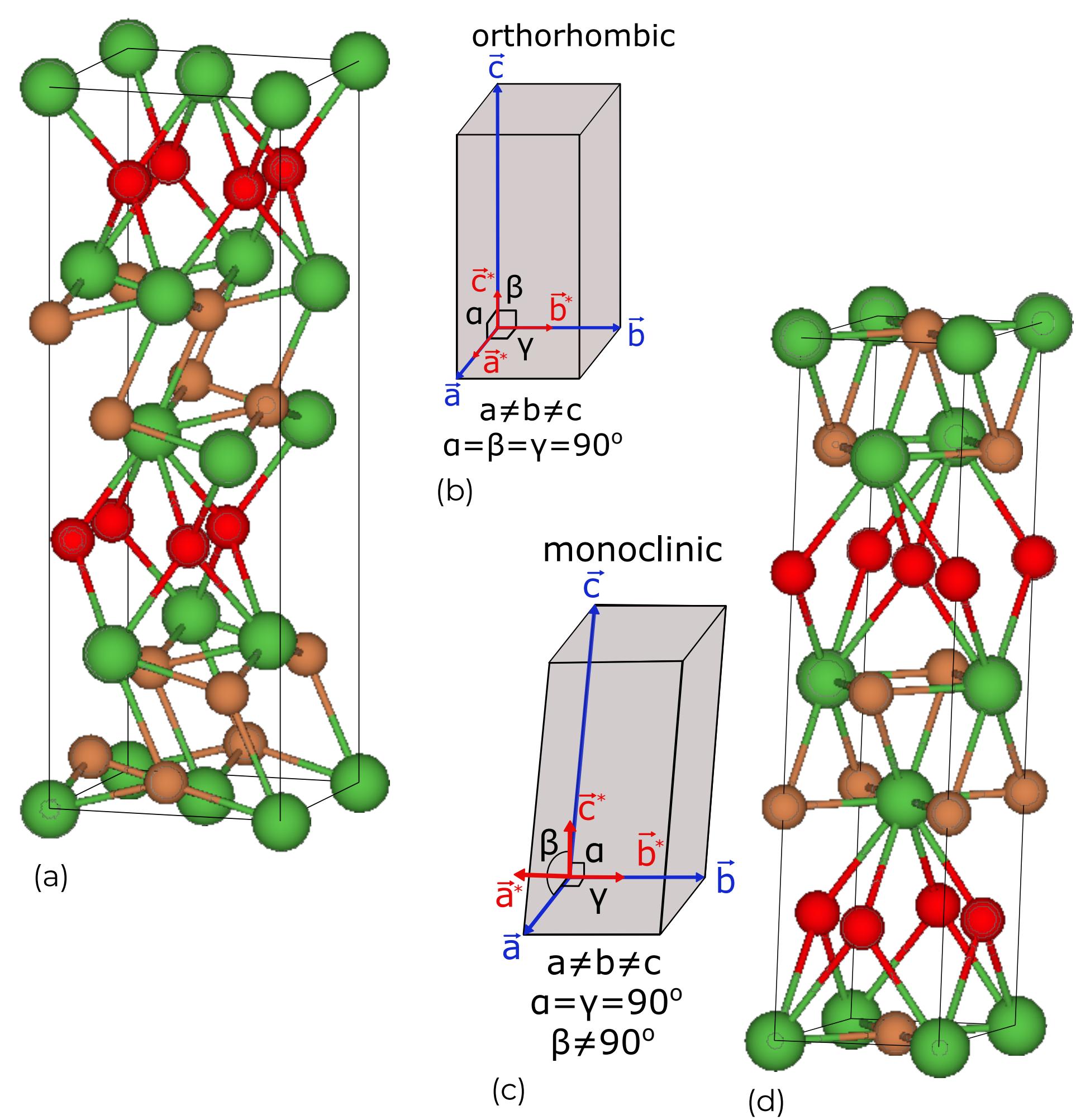


Fig. 1: a-b) Unit cell of bulk LaSb₂, shown with cartoon of orthorhombic geometry.¹ c-d) Unit cell of thin-film LaSb₂, shown with cartoon of monoclinic geometry.¹

Experimental Methodology

- LaSb₂ thin film specimens were synthesized using Molecular-beam epitaxy (MBE) on MgO substrates. In-situ Reflection high-energy electron diffraction (RHEED) measurements confirmed the crystalline order of the films, shown in Fig. 2a,b below.
- Lattice constant mismatch between the substrate and the in-plane lattice constants of the bulk crystal ($a = 6.31 \text{ \AA}$, $b = 6.18 \text{ \AA}$, $c = 18.56 \text{ \AA}$) provided an environment in which the LaSb₂ films grew under epitaxial strain.
- The crystal structure of the synthesized thin films were experimentally analyzed using Reciprocal space mapping (RSM) via X-ray Diffraction (XRD).

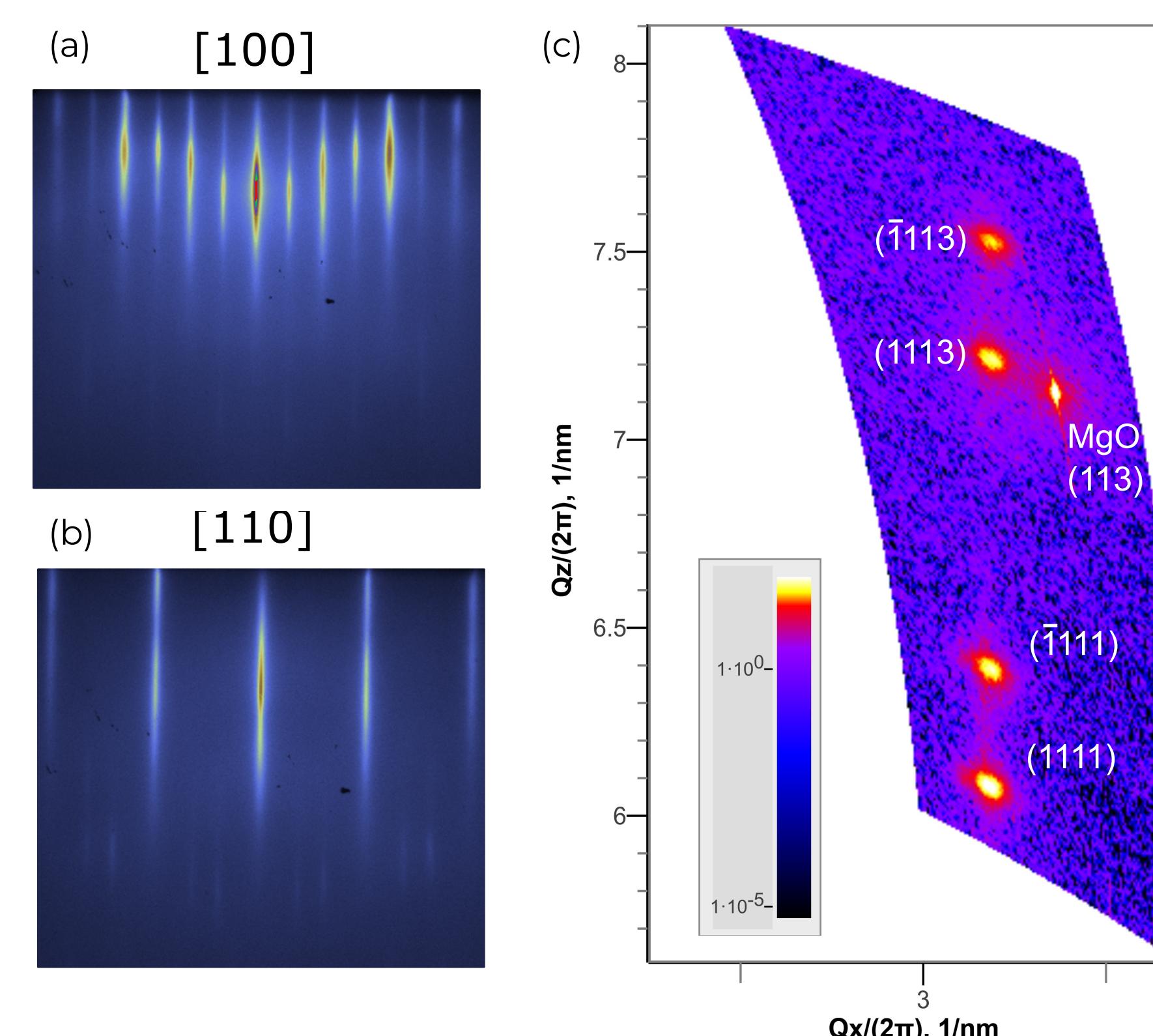


Figure 2: a-b) RHEED images for the [100] and [110] planes of thin-film LaSb₂.¹ c) Thin-film RSM showing 'twinned' intensity spots.¹

- The resulting diffraction intensity map in reciprocal space, shown in Fig. 2a above, was compared to the RSM data of the bulk LaSb₂ crystal and were found to not match, indicating that the thin-film LaSb₂ crystallized into a different space group.
- "Twinned" intensities in the RSM of the thin-film LaSb₂, shown in Fig. 2a above, indicated that the crystal structure had different in-plane lattice constants ($a = 4.56 \text{ \AA}$, $b = 4.48 \text{ \AA}$).
- From the RSM XRD data, the crystal structure of the thin-films were measured to have lattice constants $a = 4.56 \text{ \AA}$, $b = 4.48 \text{ \AA}$, $c = 17.71 \text{ \AA}$, and be monoclinic with an angle $\beta = 86.3^\circ$, shown in Fig. 1d (left). Ab-initio calculations were performed to find the crystal structure and space group of the thin-film LaSb₂.

Ab Initio Calculations

Density Functional Theory calculations were performed using the Vienna Ab-Initio Simulation Package (VASP).



- Starting from the potential *RSb*₂ parent structures, potential structures for thin-film LaSb₂ were generated by three methods: i) global deformation of the 'parent' unit cell, ii) relative displacement of atomic layers or multi-layer blocks within the 'parent' unit cell or iii) a combination of i) and ii).
- Proposed structures were relaxed first under epitaxial conditions and their ground state energies compared to the bulk. If preferential, the structure was freely relaxed and the final ground-state energy, geometry and calculated XRD pattern was compared against the experimental results.
- The ground structure of thin-film LaSb₂ was found to be the Yb-type *RSb*₂ monoclinic structure, in SG #12 A 2/m.
- Further analysis of the Yb-type *RSb*₂ monoclinic, the parent Sm-type *RSb*₂ and the parent Yb-type *RSb*₂ structures showed that a common 'unit block' could be isolated.
- By stacking unit blocks with relative displacement vectors between them, these three structures could be recreated. Fig. 3b below.

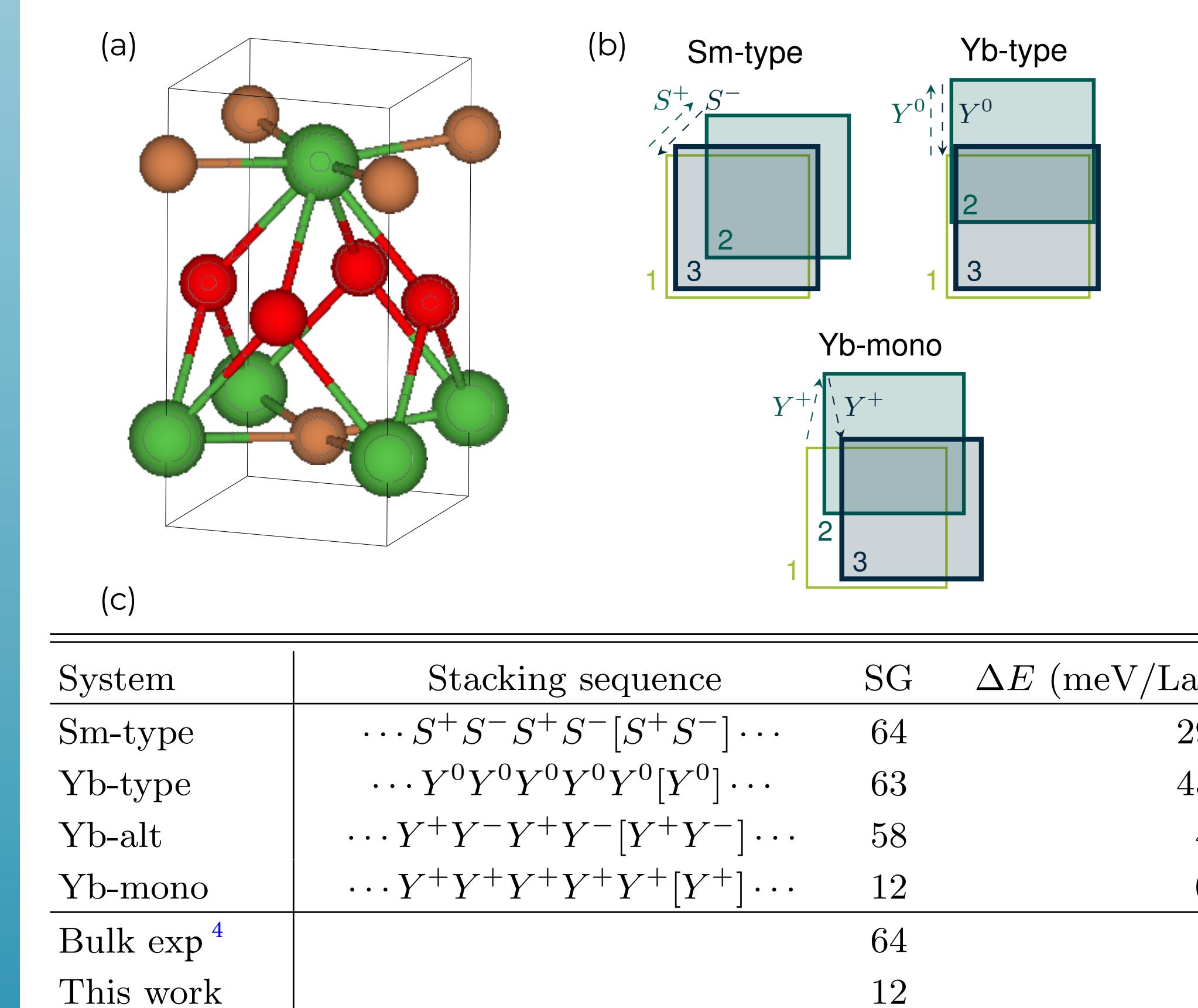


Fig. 3: a) Unit block of related structures, with C4 symmetry. b) Stacking configuration map of Sm-type, Yb-type and the new Yb-type monoclinic structure.¹ c) Table of ground state energies of stacking configurations.¹

Results

- The experimental and computational results show that LaSb₂ forms a novel monoclinic ground state in epitaxially constrained thin-films.
- The square-net of Sb²⁺ ions and superconductivity at ambient conditions remain in the thin-films. Transport measurements show that both carrier mobility and density are changed w.r.t the bulk crystal, with the critical temperature for superconductivity T_c being higher in the thin-films than in the bulk.¹
- Further calculations on the relative displacement of unit blocks show a hitherto unknown potential structure, dubbed Yb-alt, which is preferential to both the Sm- and Yb-type *RSb*₂ parent structures.
- The research presented shows the viability of tuning the coupled electronic and structural characteristics in the family of *RSb*₂ layered intermetallic systems through thin-film growth under epitaxial strain.

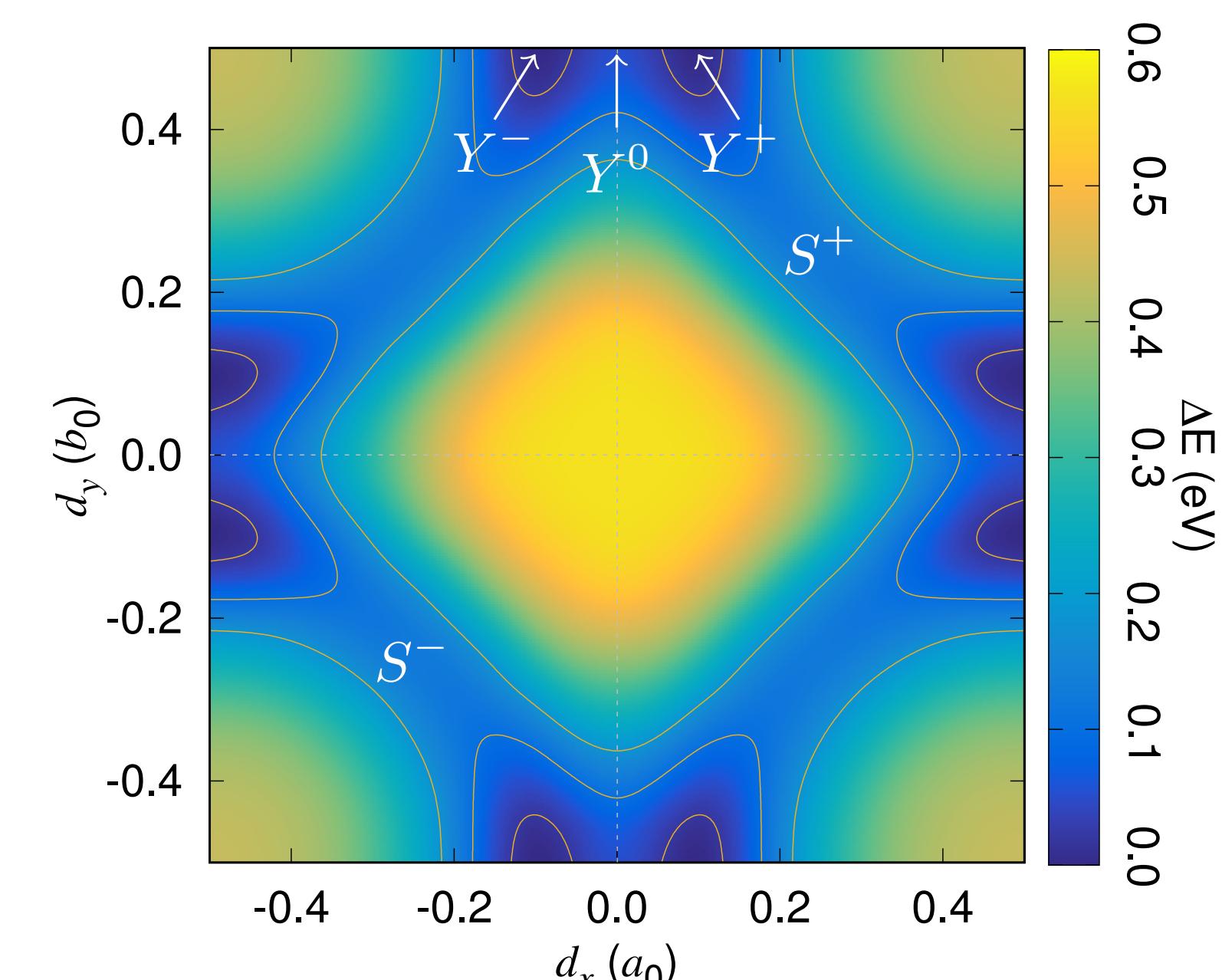


Fig. 4: Energy map w.r.t the relative displacement of two unit blocks. The S[±] and Y⁰ saddle points reflect the Sm- and Yb-type *RSb*₂ parent structures respectively.¹

References

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